On Controlling the Gamma Decay of a Nuclear Excited State

K.P. Heeg et al., Nature 590, 4021 (2021)

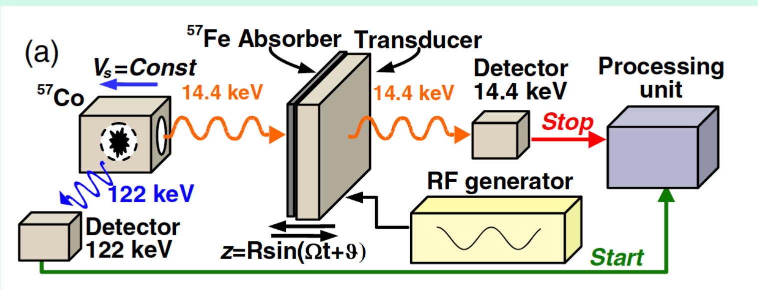
Coherent X-ray-optical control of nuclear excitons

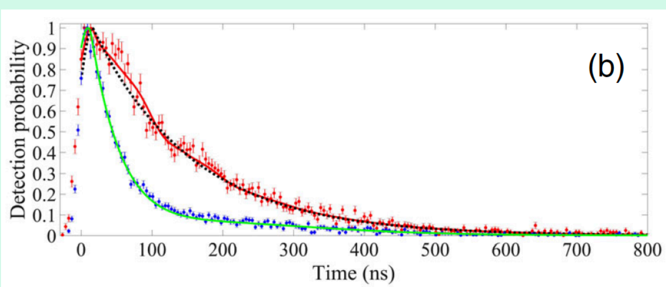
and related works

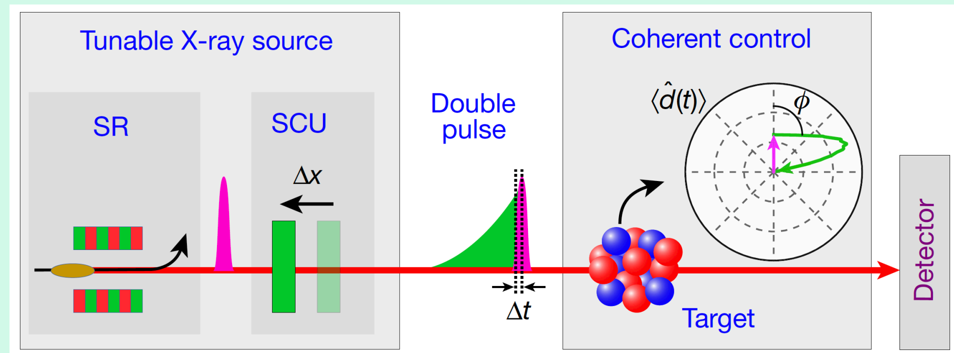
Super-radiance

Storage of nuclear excitation energy

Acoustically induced transparency

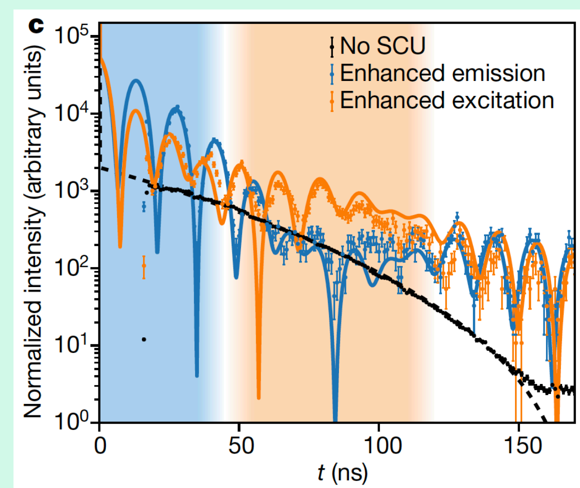






Mössbauer spectroscopy

zeptosecond stability



Presenter: A. Tamii

Kakuri-1 Benkyokai on June 3&10, 2022, RCNP, Osaka University

Article

Coherent X-ray-optical control of nuclear excitations

K.P. Heeg et al., Nature 590, 401 (2021)

Abstract:

Coherent control of quantum dynamics is key to a multitude of fundamental studies and applications. In the visible or longer-wavelength domains, near-resonant light fields have become the primary tool with which to control electron dynamics. Recently, coherent control in the extreme-ultraviolet range was demonstrated, with a few atto-sec temporal resolution of the phase control. At hard-X-ray energies (above 5–10 keV), Mössbauer nuclei feature narrow nuclear resonances due to their recoilless absorption and emission of light, and spectroscopy of these resonances is widely used to study the magnetic, structural and dynamical properties of matter. It has been shown that the power and scope of Mössbauer spectroscopy can be greatly improved using various control techniques. However, coherent control of atomic nuclei using suitably shaped near-resonant X-ray fields remains an open challenge. Here we demonstrate such control, and use the tunable phase between two X-ray pulses to switch the nuclear exciton dynamics between coherent enhanced excitation and coherent enhanced emission. We present a method of shaping single pulses delivered by state-of-the-art X-ray facilities into tunable double pulses, and demonstrate a temporal stability of the phase control on the few zepto-sec timescale. Our results unlock coherent optical control for nuclei, and pave the way for nuclear Ramsey spectroscopy and spin-echo-like techniques, which should not only advance nuclear quantum optics, but also help to realize X-ray clocks and frequency standards. In the long term, we envision time-resolved studies of nuclear out-of-equilibrium dynamics, which is a long-standing challenge in Mössbauer science.

atto-sec = 10^{-18} s
zepto-sec = 10^{-21} s

Contents

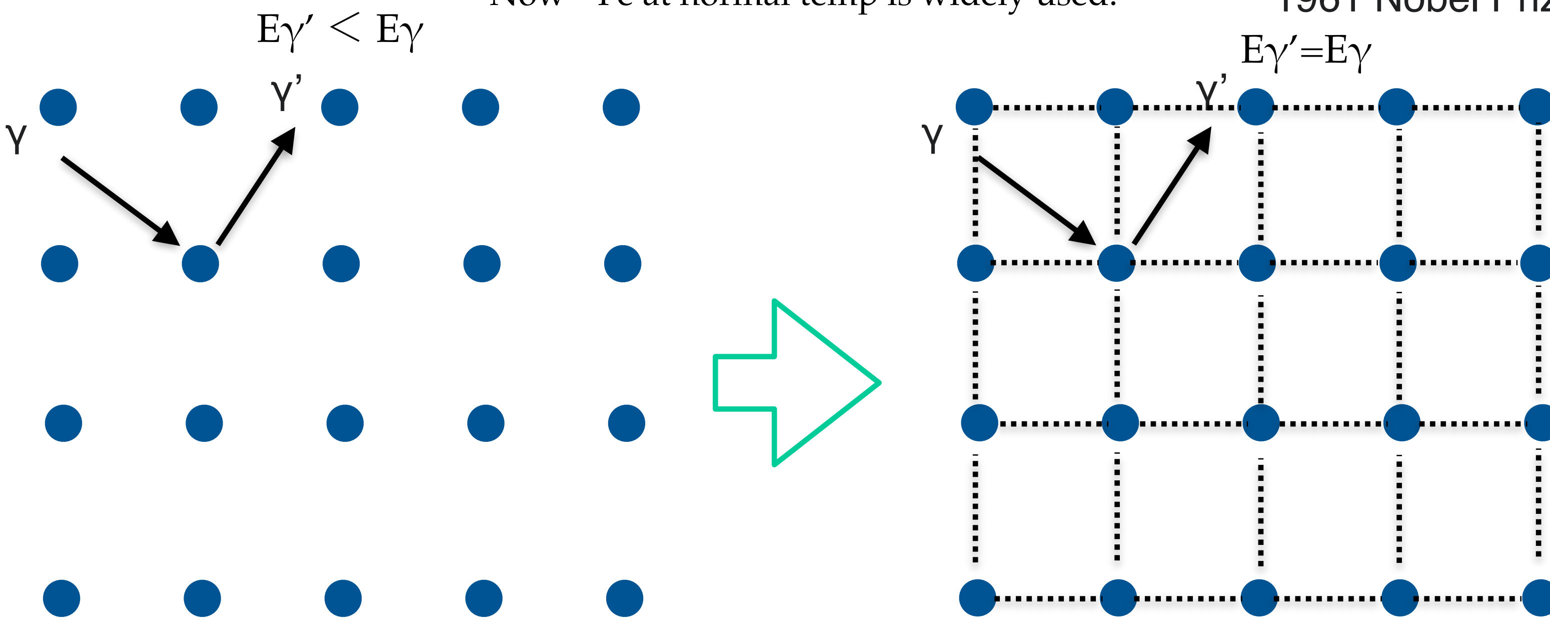
Mössbauer Effect

^{191}Ir at low temp.

Now ^{57}Fe at normal temp is widely used.

1958 Rudolf Mössbauer

1961 Nobel Prize



Usually the γ′ energy is smaller than γ due to the recoil of the nucleus.

Nuclear **recoil energy is reduced due to lattice structure of metal/crystal**, in some case, with reduction of heat motion at low-energy.

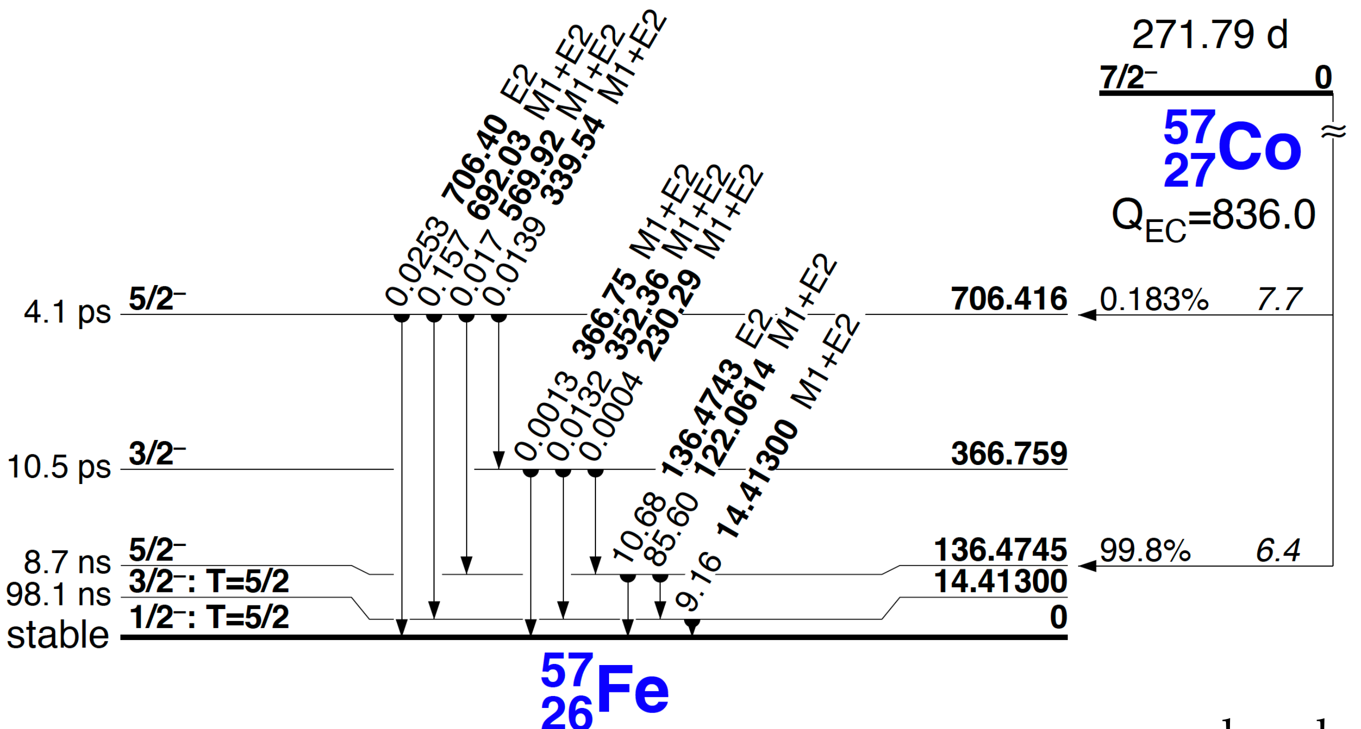
- The emitted γ-can be captured by another nucleus.
- **An ensemble of nuclei becomes a coupled system for coherent γ-excitations/emissions**

Recoil energy smaller than phonon.

→ **Mössbauer spectroscopy** for **very precise measurements** ($\sim 10^{-12}$), e.g. verification experiment of general relativity

→ **Coherent control of nuclear quantum systems**

^{57}Fe



abundance: 2.12%

3/2- state at E=14.4 keV

$T_{1/2}$=98.1 ns

τ = 141.5 ns

$\Gamma = 4.65\times10^{-9}$ eV (γ in this article)

$E/\Gamma = 3.1\times10^{12}$

$\lambda = 0.86$Å

Levels:

0, 1/2⁻, stable,
[ACDEFGHIJKLMNORSTUVX],
μ=+0.09044 *7*, T=5/2

14.41300 *15*, 3/2⁻, 98.1 *3* ns,
[ACDEFGHIJKLMNORSTVX],
μ=−0.1549 *2*, Q=+0.082 *8*, T=5/2
γ_0 ***14.41300*** *15* ($\dagger_\gamma$100) M1+E2: δ=0.00219 *17*

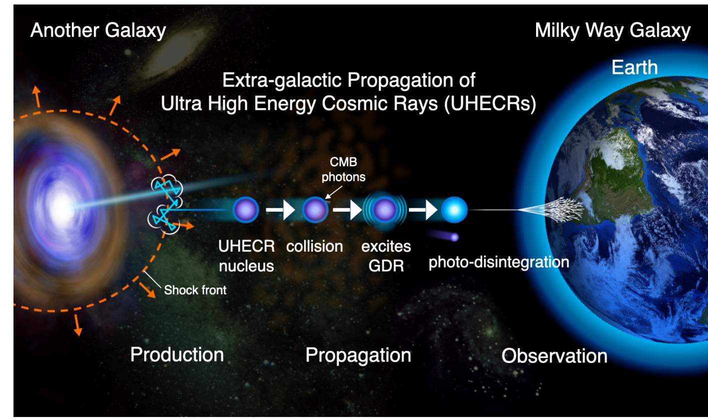
My personal Interest

PANDORA Project

Photo-Absorption of Nuclei and Decay Observation for Reactions in Astrophysics

Photo-excitation and **Decay** up to A~56

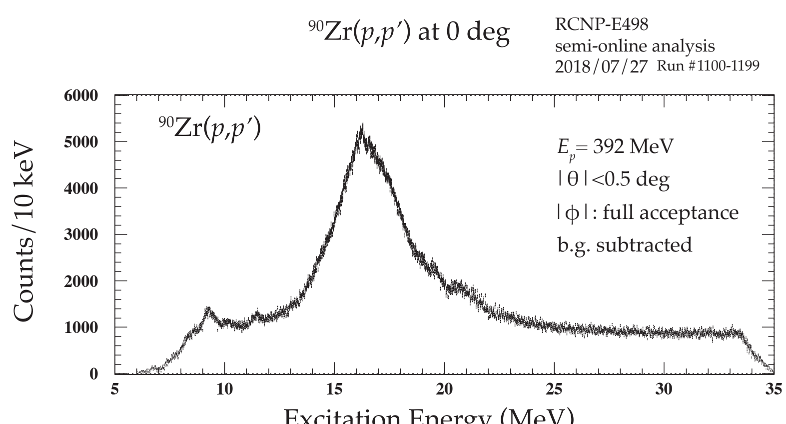
- E1 excitation strength distribution
- n, p, α, γ decay branching ratios
- from light to A~56 for stable nuclei

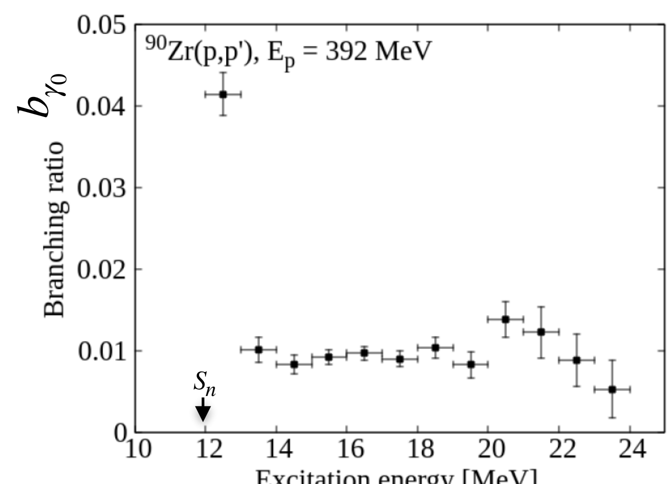


Gamma Decay of IVGDR

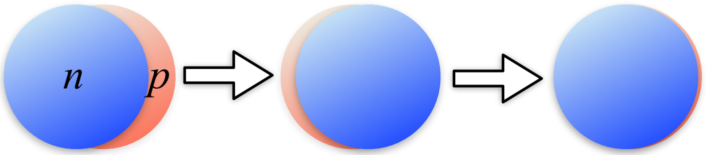
Origin of **width**, damping, spreading

Pre-equilibrium vs Compound





Spreading of IVGDR



Damping of IVGDR

IVGDR

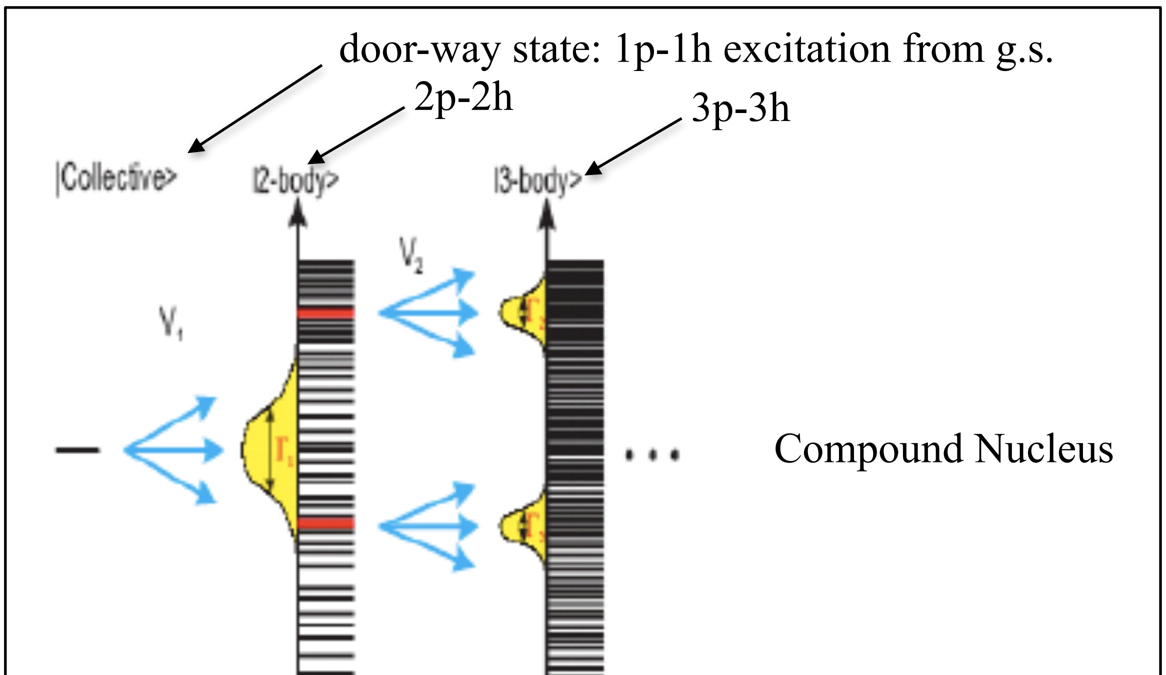
macroscopically: relative dipole oscillation between *n* and *p*

microscopically: **coherent superposition** of 1p-1h excitations

Spreading

macroscopically: **viscosity** between *n* and *p* liquids

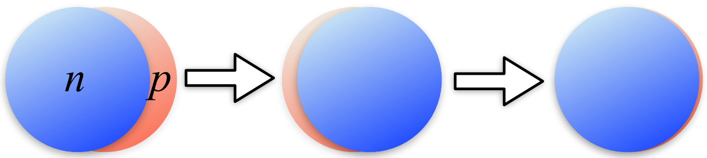
microscopically: **loss of coherence** of the 1p-1h excitations



out of coherence

Bracco, Lanza, and AT, PPNP'19

Spreading of IVGDR



Damping of IVGDR

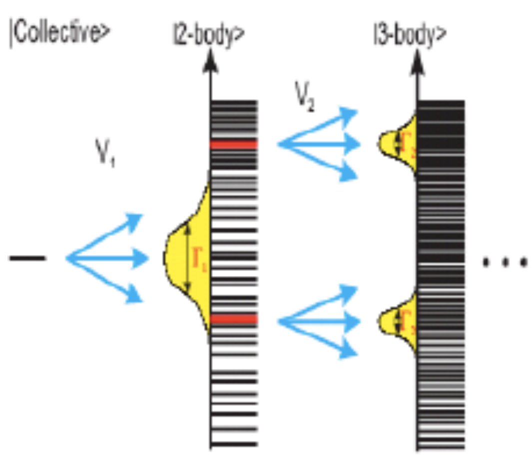
Spreading width:

W.F. of an excited state

doorway collective state excited from the g.s.

$$|\psi>_i = |\mathrm{Collective}>_i + \Sigma|2p2h>_i + \ldots + \Sigma|CN>_i$$

damping with a loss of coherence



- **Pre-equilibrium state** still keeps the information on the ***phase***. The decay depends on the *phase*.
- Coherent excitation of 1p-1h states in IVGDR ⇔ coherent excitation of Mössbauer nuclei
  There might be analogous phenomena specific in Mössbauer physics

Introduction

Coherent control:

- quantum dynamics by light
- phenomena relevant to coherence and interference

→ requires shaping of light pulses

exciton dynamics of an ensemble of nuclei

e.g., exciton (polariton) propagation or radiation trapping

- by **magnetic switching**
- interference between different scattering pathways by **mechanical motion of resonant absorbers**

"We demonstrate the coherent control of the dynamics of Mössbauer nuclei using X-ray light."

Introduction

Controlling the relative phase of the second pulse

→ Switching of the subsequent target dynamics between

- coherent enhanced excitation
- coherent enhanced emission

of the nuclear exciton

We detect **time-dependent magnitude and phase** of the spatially averaged **transition dipole moment**

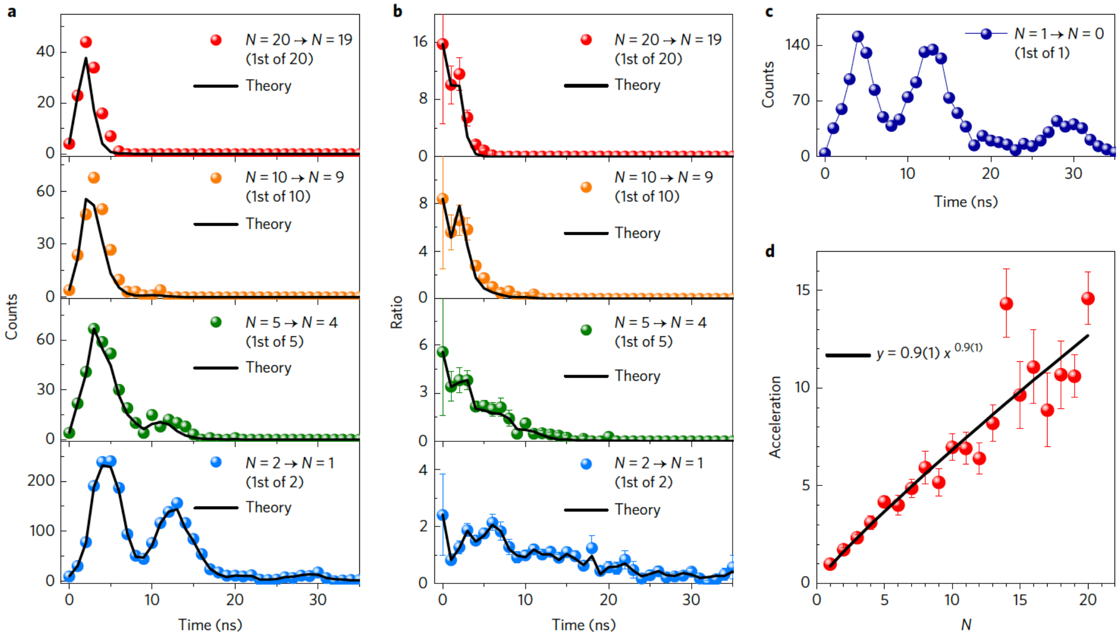
We demonstrate **a few-zeptosecond temporal phase stability** related works

Superradiance R.H. Dicke PR93, 99 (1954)

A.I. Chumakov et al., Nature Phys. 14, 261 (2018)

$^{57}FeBO_3$ single crystal

$\tau =$ 141.5 ns



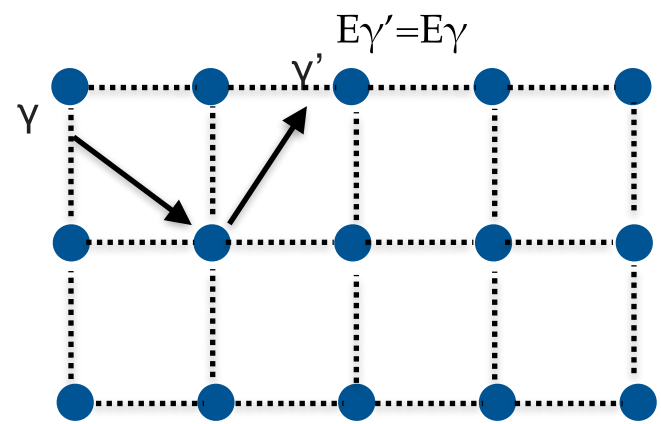


Fig. 3 | Initial decay rate. a, The increase in the initial decay rate for the transitions from N to $N-1$ excited states is revealed by the accelerated decay of the first out of N detected photon, $P_N^1(t)$. **b**, The ratios $P_N^1(t)/P_1^1(t)$ of these data to the single-photon decay $P_1^1(t)$ also illustrate this increase. **c**, Single-photon decay. **d**, Estimated acceleration rates $(P_N^1/P_1^1)|_{t\to 0}$ (see Supplementary Information for details). The solid lines in **a**,**b** are the calculations according to equation (2). The solid line in **d** is the power fit. The error bars show the standard deviations related to the numbers of counts per channel in the raw data (**b**) and to the uncertainty of the linear fit to $P_N^1(t)/P_1^1(t)$ (Supplementary Information).

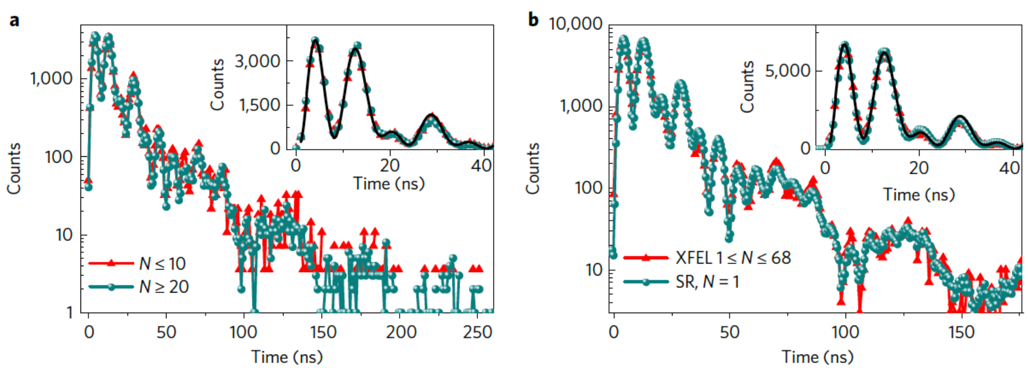
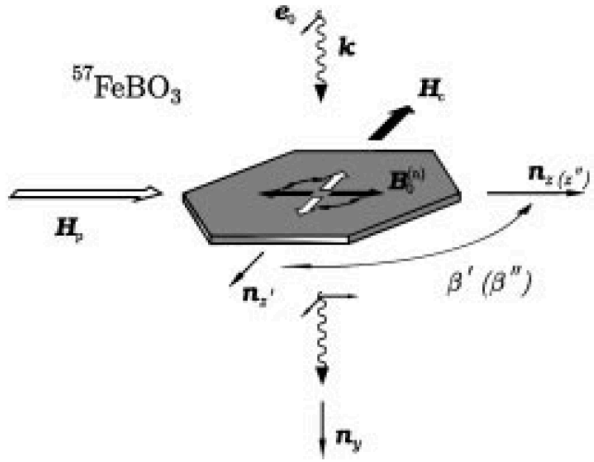
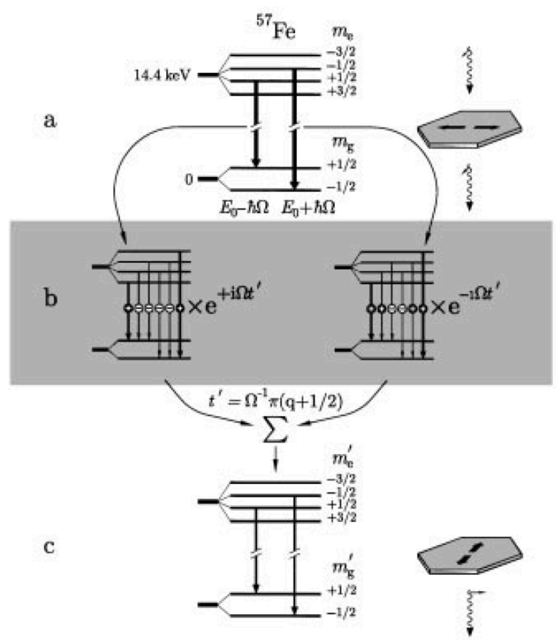


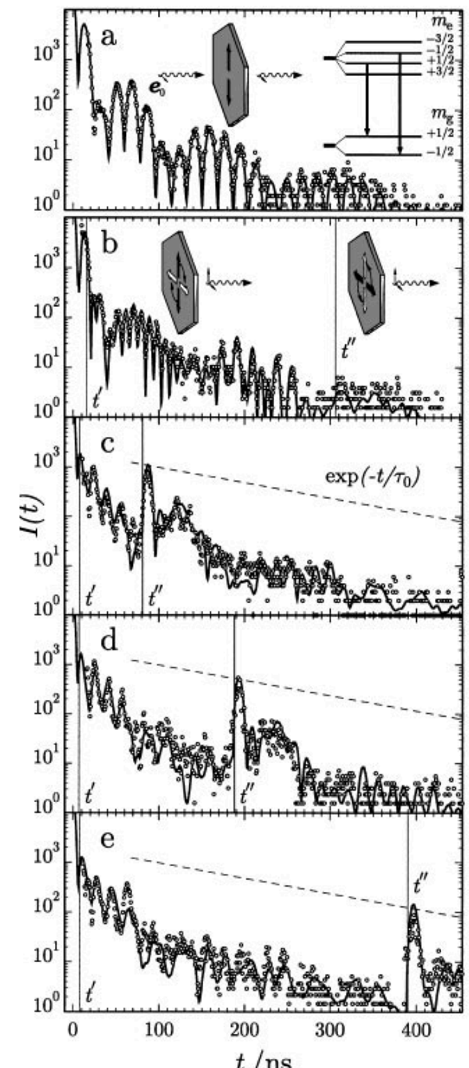
Fig. 4 | Emission of the total energy. a, The decay including all photons for the events where either $N \leq 10$ or $N \geq 20$ photons were detected. These are the same within statistics. **b**, The decay over all photons measured at the XFEL with multiphoton events is the same as what is measured at a synchrotron radiation beamline with single-photon events. The solid line in the insets illustrates theory[22] that shows that the decay of the nuclear ensemble can be calculated with great accuracy for single-photon excitations.

Magnetic Switching

Yu.V. Shaved’ko et al., PRL77, 3232 (1996).

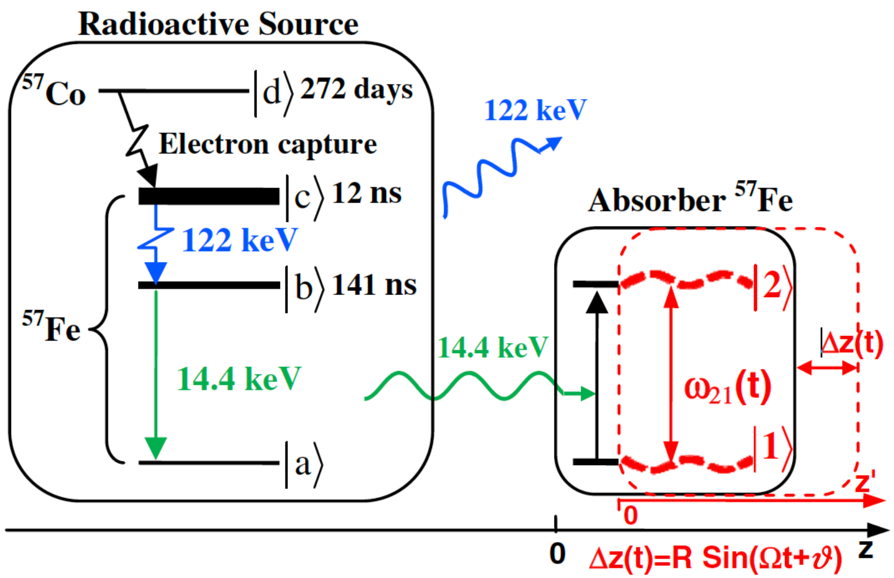


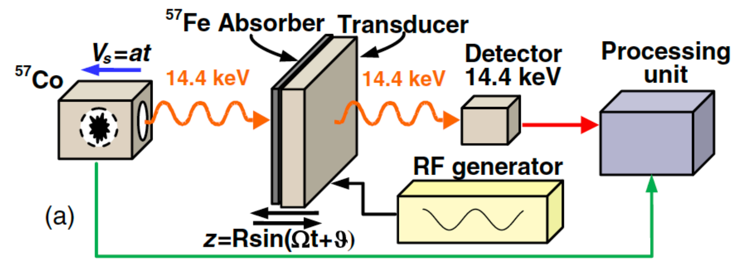


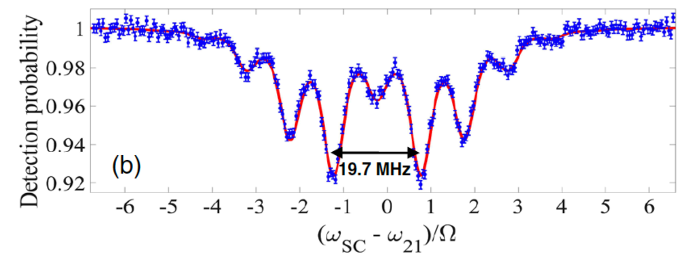


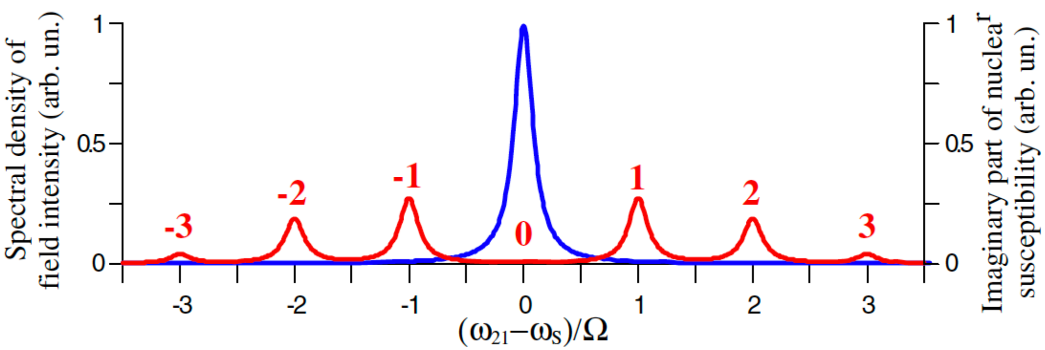
Acoustically Induced Transparency

Y.V. Radeonychev et al., PRL124, 163602 (2020)









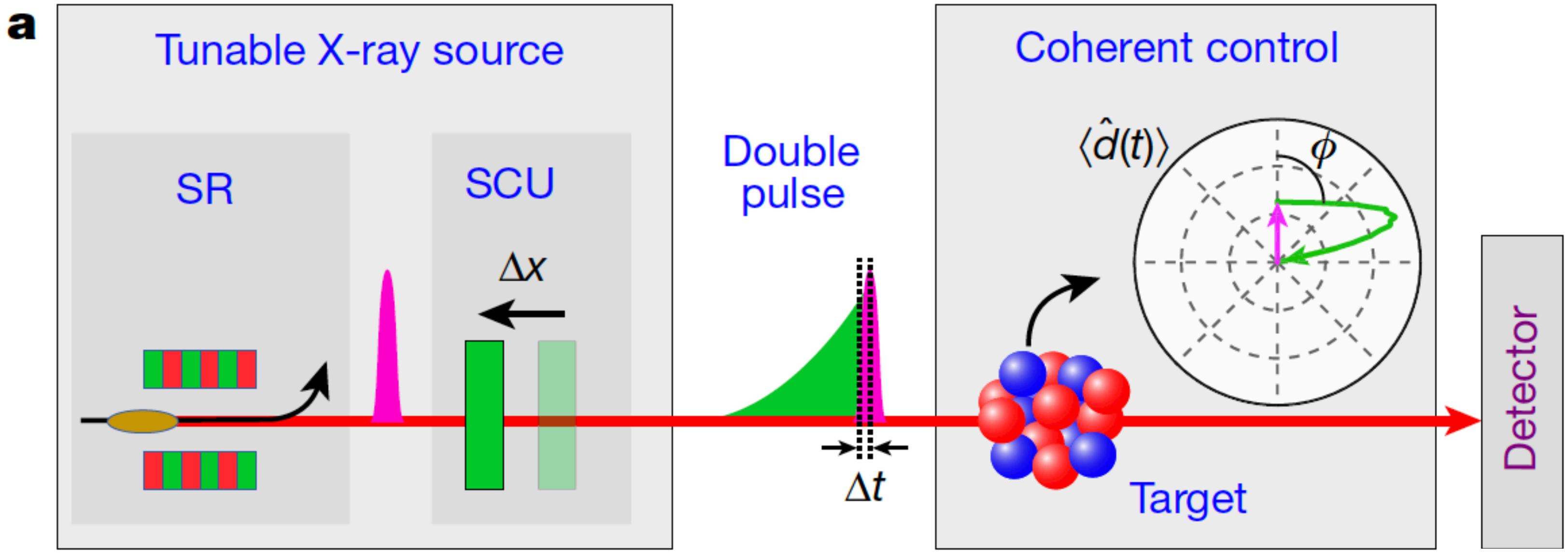
$$J_0(2\pi R_i/\lambda) \approx 0$$

$$R = R_i, \quad \text{where } R_1 \approx 0.38\lambda, R_2 \approx 0.88\lambda, R_3 \approx 1.37\lambda, \ldots, \quad (1)$$

this work

Experimental Setup

@European Synchrotron Radiation Facility (ESRF) in Grenoble



photon by photon

SCU: Split and Control Unit, α-iron foil 2μm (^{57}Fe enriched)

<u>excitation pulse</u>: non-delayed fraction of the X-ray (short)

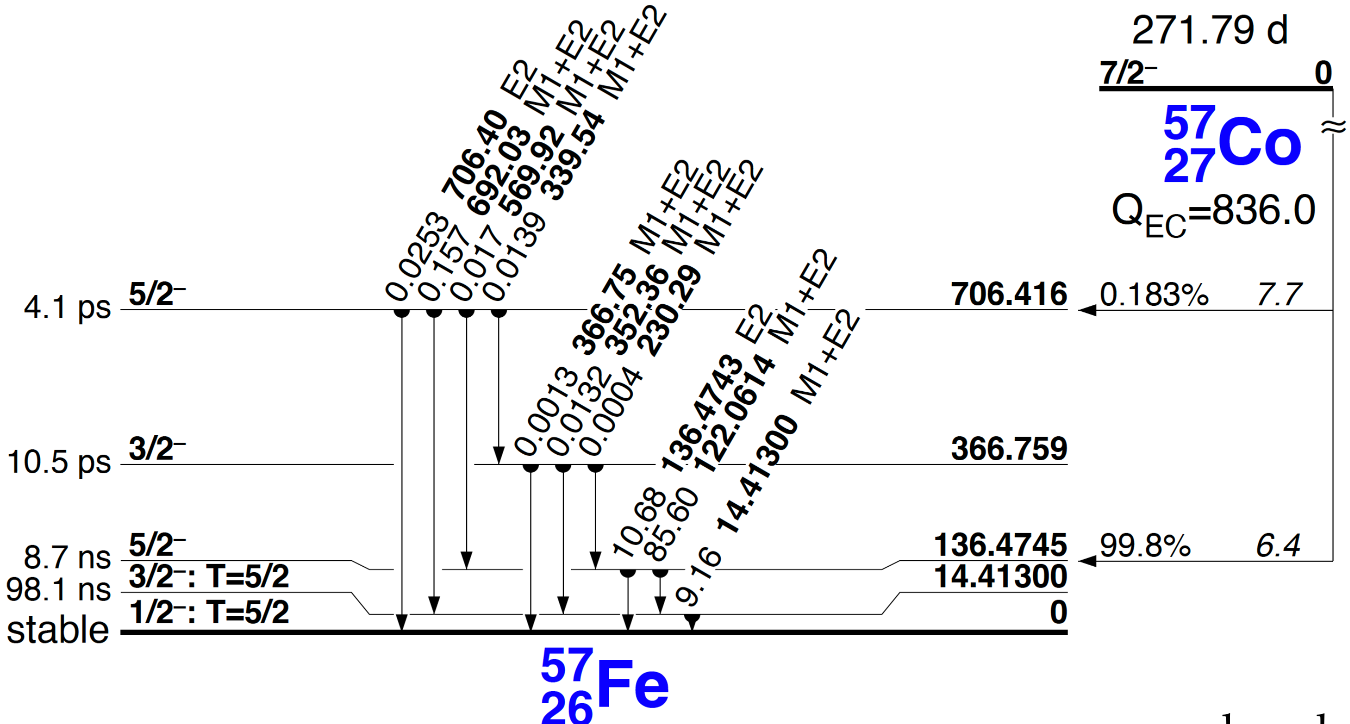
<u>control pulse</u>: delay by absorption/emission by SCU

+ phase control, $\exp\{ikx(t)\}$, by mechanical movement of SCU

narrow width ~ neV

Target: Stainless Steal 1μm (57Fe: 95% enriched)

^{57}Fe



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3/2⁻ state at E=14.4 keV

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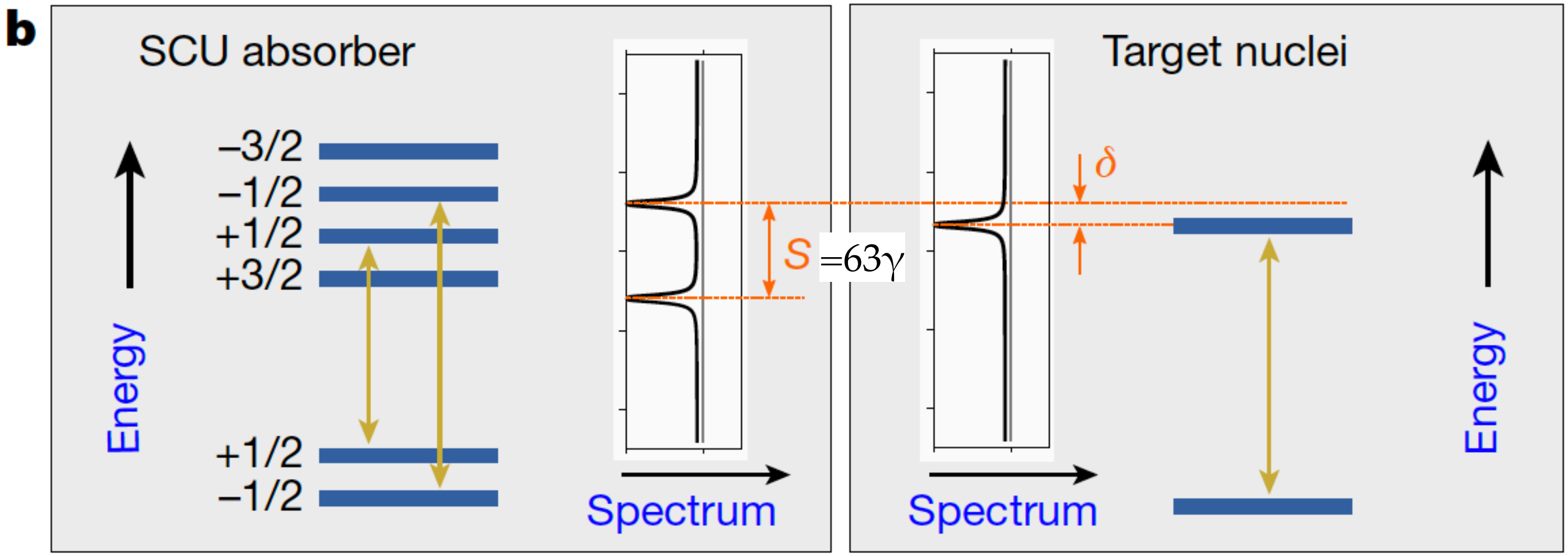
Experimental Setup



A weak external magnetic field

→ drive only the two Δm=0 transitions

Doppler shift → energy scanning

relative phase: 0	coherent enhance excitation
π	coherent enhance emission

Experiment

Detection of photon at the forward direction (~0^{o}):

photon by photon (Event by Event)

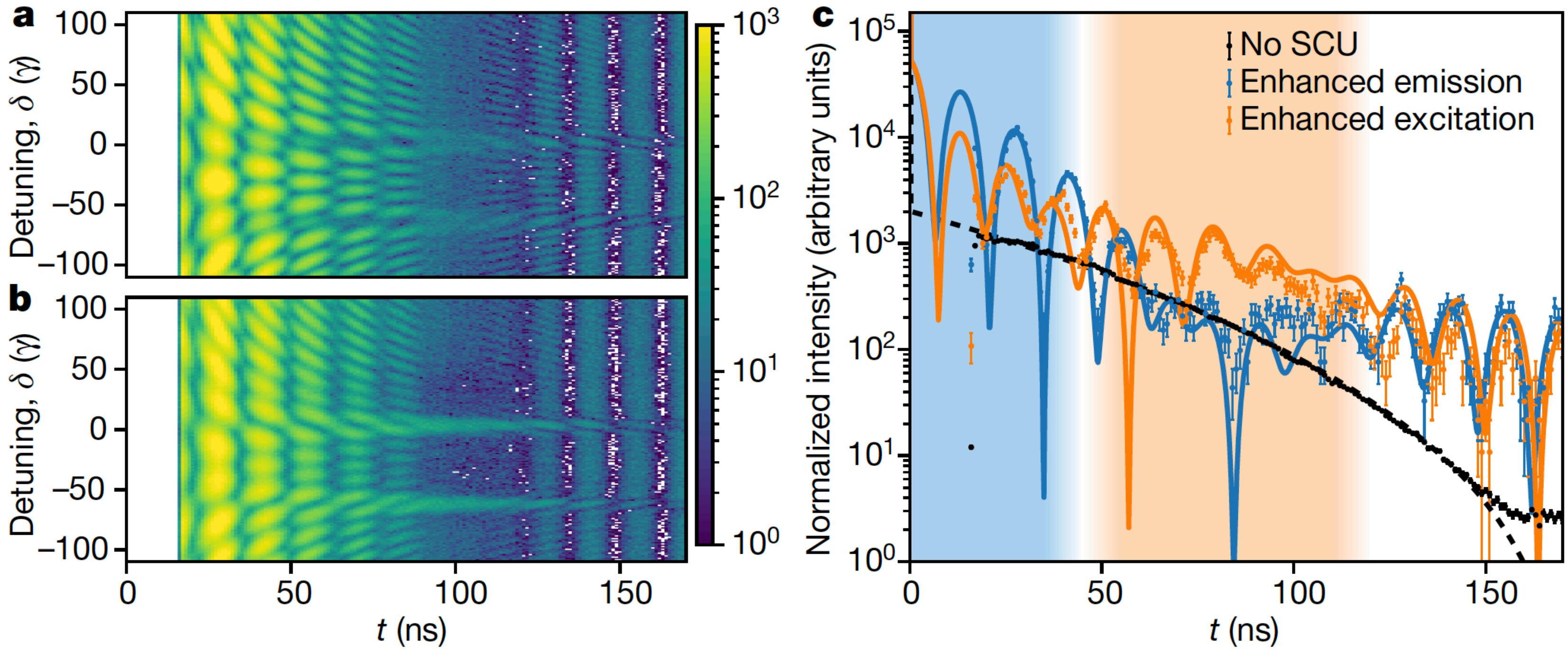
amplitude proportional to

the spatial average over the nuclear magnetic transition dipole moment:

$$\left\langle \hat{d}(t, \delta) \right\rangle$$

Result

at $\delta=0$



a: Enhanced excitation $\Delta\phi=0$

solid lines: theory curves

b: Enhanced excitation $\Delta\phi=\pi$

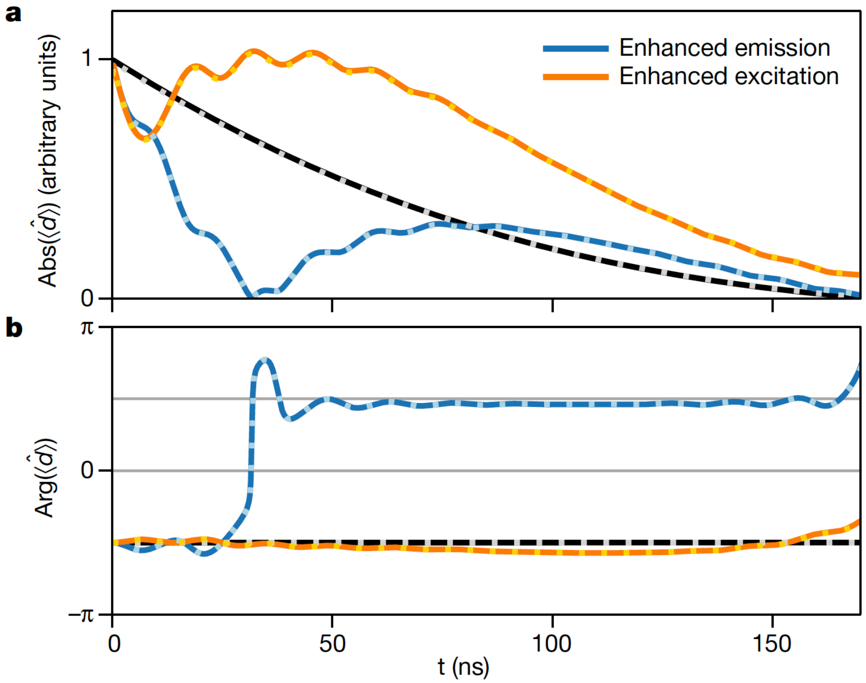
c:

blue region: enhanced emission is higher (superradiance)

orange region: enhance excitation is higher (increased excitation)

oscillation: quantum beat between two lines from SCU

Result



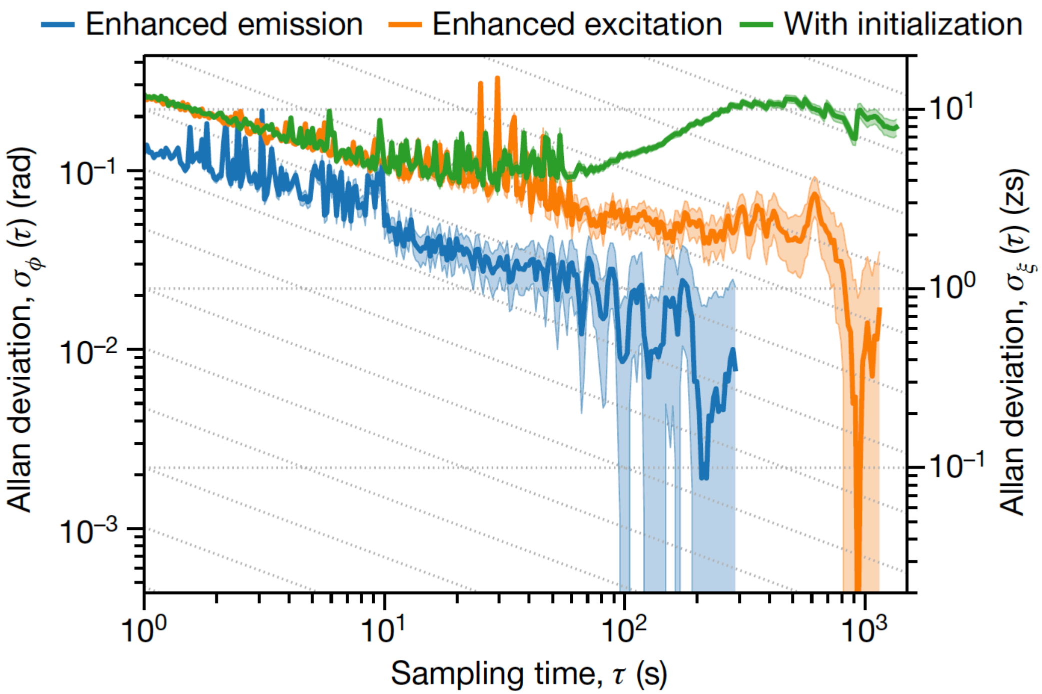
Amplitude

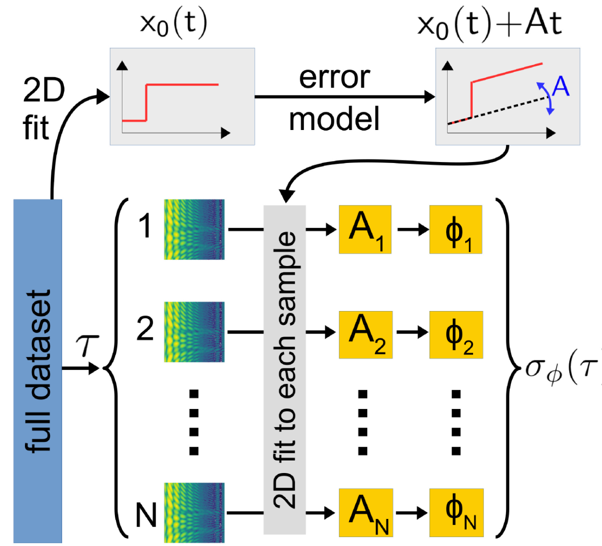
Phase

$$\left\langle \hat{d}(t,\delta) \right\rangle$$

Reconstructed the spatial average of the nuclear magnetic transition dipole moment deduced by a fit to the experimental data.

Results





The phase stability

τ: interval of the samples

The deviation decreases as τ increases since the noises are more effectively smoothed-out.

The minimum, intrinsic phase-stability, is not seen (the phase-stability is masked by the noise of the detection system).

stability: 40 mr, a few zepto-sec, best reported (two-orders better)

Future possibilities

- Addition of X-ray pulse storing by magnetic switching
- Stronger X-ray by a free-electron laser system
- Engineering complex quantum states
- Exploring time-dependent phenomena with nuclei
- Nuclear out-of equilibrium dynamics

Remarks

- Strong g.s. γ-decay of IVGDR

  large coupling to the initial state

  Connection to super-radiance?
- γ-emission angular distribution characteristic to super-radiance?
- Characteristic feature in high-field or in multi-photon excitation?

- Coherent excitation of IVGDR in an ensemble of nuclei?

  conditions would not be fulfilled

  - nuclear recoil energy ~10 keV
    (is there a characteristic direction?)
  - γ-wavelength is much smaller than the distance of nuclei
    (coherence at θ< 1mr ?)
  - coupling (photo-abs. c.s.) is much weaker

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Backups